The influence of four-particle correlations on the parametric polariton amplification

Salvatore Savasta[*], **Omar Di Stefano,** and **Raffaello Girlanda**

Istituto Nazionale per la Fisica della Materia INFM and Dipartimento di Fisica della Materia e Tecnologie Fisiche Avanzate, Università di Messina Salita Sperone 31, I-98166 Messina, Italy

We present a microscopic theory of parametric polariton amplification in semiconductor microcavities including the influence of exciton–exciton correlations beyond mean-field. The complexity induced by the Coulomb interaction between electrons determines the noninstantaneous character of exciton–exciton collisions. We find that the observed great enhancement of gain when increasing the polariton splitting origins from the unique interplay of the noninstantaneous nature of exciton–exciton interactions with the strong coupling regime giving rise to polaritons.

Polaritons in semiconductor microcavities (SMCs) are two-dimensional eigenstates resulting from the strong resonant coupling between cavity-photon modes and two-dimensional excitons in embedded quantum wells (QWs) [1]. The dynamics and hence the resulting energy bands of these mixed quasi-particles are highly distorted respect to those of bare excitons and cavity photons. The exciton-photon coupling rate V determines the splitting ($2V$) between the two polariton energy bands. The huge phase-coherent amplification of cavity-polaritons recently demonstrated [2] is attracting great attention [3–8, 10] also for the possible future development of all-optical devices [11]. More recently it has been shown that, increasing the exciton-photon coupling, greatly increases amplification [11] (gains up to 5,000 have been observed), allowing the design of several possible microscopic devices approaching room temperature operation.

We present a microscopic theory of parametric polariton amplification in semiconductor microcavities including the influence of exciton–exciton correlations beyond mean-field. We find that the strong coupling of excitons with cavity-photons alters the excitonic dynamics during exciton–exciton collisions, producing a modification of the effective scattering rates. Since the dynamics of the coupled excitons and photons is determined by the exciton-photon coupling rate, its variation is able to affect significantly exciton–exciton collisions and hence the amplification process. Thus we show that the possibility of observing very large gains is a consequence of the coherent control that the exciton–photon interaction exerts on the not instantaneous exciton–exciton collisions. This control is able to produce almost decoherence-free exciton–exciton collisions of great importance for the development of future all-optical devices.

Coherent amplification of polaritons requires a coupling mechanism able to transfer polaritons from a reservoir (in this case provided by polaritons resonantly excited by a pump laser pulse on the lower polariton dispersion) to the signal mode, while conserving energy and momentum. As shown by the microscopic theory of parametric polariton amplification [8], polaritons of different modes are coupled

[*] Corresponding author: e-mail: Savasta@unime.it

via their excitonic content, the coupling being provided mainly by the Coulomb interaction between excitons (also the anharmonic part of the exciton-photon interaction contributes). The microscopic theory of parametric polariton amplification [8, 9] puts forward the parametric and coherent nature of this process. It also shows that the resulting Coulomb coupling strength is given by the exciton–exciton (XX) scattering rate [18] $V_{XX} \simeq 1.52 E_b a_0^2$ (E_b is the exciton binding energy and a_0 is the exciton Bohr radius) times the exciton fraction of the interacting polariton modes. This effective polariton-polariton interaction scatters a pair of pump polaritons into the lowest-energy state and into a higher-energy state (usually known as the idler mode) (energy and momentum conservation requires $2\omega_k = \omega_0 + \omega_{2k}$, where ω_k is the energy of a pump polariton injected with an in-plane wavevector $\boldsymbol{k}$). The scattering process is stimulated by a weak signal beam injected perpendicular to the cavity ($\boldsymbol{k} = 0$) that is thus greatly amplified. It has recently been shown that, increasing the exciton-photon coupling V by inserting a large number of QWs into the cavity, greatly increases amplification [11]. According to mean-field descriptions, the gain is limited only by the pump depletion and (when injecting a small signal beam) can thus reach very high values also for small values of V (unless some additional saturation mechanism is introduced). Moreover according to these descriptions, inserting more QWs just lowers the polariton-polariton interaction, without changing the structure of pertinent equations (the exciton and photon contents of the 3 interacting modes result to be independent on V). From this observation and from a direct inspection of the pertinent equations, it descends that, increasing the exciton-photon coupling, should not greatly increase the amplification of a weak signal beam in contrast with the spectacular experimental observations [11].

The mean-field theory of the optical response of electron-hole pairs assumes that collisions are instantaneous and only the mean free time between collisions matters. Although the mean-field theory has enjoyed considerable success [9, 14], recent studies revealed that collisions between excitons are more complex (see [13] and references herein cited). They involve multiparticle (at least 4-particle) entangled states. This complexity induced by the Coulomb interaction between electrons, determines the finite duration of collisions [13, 15–19]. We find that the strong coupling of excitons with cavity-photons, giving rise to polaritons, alters the excitonic dynamics during XX collisions, producing a modification of the effective scattering rates. As a consequence, a variation of the exciton-photon coupling rate V can affect significantly XX collisions, and hence, the amplification process. To analyze the influence of the finite duration of XX interactions on the parametric polariton amplification, we use a microscopic description where the hierarchy of higher order density matrices has been closed by invoking the dynamics controlled truncation (DCT) scheme [20] in the coherent limit. The intracavity light-field is described within the quasi-mode scheme [16]. The time evolution of the coupled exciton ($P_{\boldsymbol{k}}$) and photon waves ($E_{\boldsymbol{k}}$) including finite duration of XX collisions [12, 16] can be described by the following set of coupled equations,

$$\begin{aligned} \frac{\partial}{\partial t} E_{\boldsymbol{k}} &= -(\gamma_c + i\omega_k^c)\, E_{\boldsymbol{k}} + iVP_{\boldsymbol{k}} + t_c E_{\boldsymbol{k}}^{\text{in}} \\ \frac{\partial}{\partial t} P_{\boldsymbol{k}} &= -(\gamma_x + i\omega_x)\, P_{\boldsymbol{k}} + iVE_{\boldsymbol{k}} - i\Omega_{\boldsymbol{k}}^{NL}, \end{aligned} \tag{1}$$

where ω_k^c, ω_x, and γ_c, γ_x are the energies and dephasing rates of cavity photons and QWs excitons respectively. $\left|E_{\boldsymbol{k}}^{\text{in}}\right|^2$ describes the input photon rate, t_c determines the beam fraction passing the cavity mirror ($t_c = \sqrt{\gamma_c}$ for a cavity with equal mirrors). The exciton photon coupling V changes with the effective number of QWs N_{eff} which depends on the number of wells inside the cavity and their spatial overlap with the cavity-mode. In particular $V = V_1 \sqrt{N_{\text{eff}}}$, where V_1 is the exciton-photon coupling for 1 QW. The relevant nonlinear source term, able to couple waves with different in-plane wave-vector $\boldsymbol{k}$, is given by $\Omega_{\boldsymbol{k}}^{NL} = (\Omega_{\boldsymbol{k}}^{\text{sat}} + \Omega_{\boldsymbol{k}}^{XX})/N_{\text{eff}}$, where the first term originates from the phase-space filling of the exciton transition,

$$\Omega_{\boldsymbol{k}}^{\text{sat}} = \frac{V}{n_{\text{sat}}} \sum_{\boldsymbol{k}'\boldsymbol{k}''} P_{\boldsymbol{q}}^{*} P_{\boldsymbol{k}''} E_{\boldsymbol{k}'}\,, \tag{2}$$

being $n_{\text{sat}} = 7/(4\pi a_0^2)$ the exciton saturation density ($\boldsymbol{k}'$, $\boldsymbol{k}''$, and $\boldsymbol{q}$) are tied by the momentum conservation relation $\boldsymbol{k}+\boldsymbol{q}=\boldsymbol{k}'+\boldsymbol{k}''$. $\Omega_{\boldsymbol{k}}^{XX}$ is the Coulomb interaction term. It dominates the coherent XX coupling and for co-circularly polarized waves can be written as

$$\Omega_{\boldsymbol{k}}^{XX} = \sum_{\boldsymbol{k}'\boldsymbol{k}''} \left(V_{XX} P^*{}_{\boldsymbol{q}}(t)\, P_{\boldsymbol{k}''}(t)\, P_{\boldsymbol{k}'}(t) - iP_{\boldsymbol{q}}^*(t) \int_{-\infty}^{t} F(t-t')\, P_{\boldsymbol{k}''}(t')\, P_{\boldsymbol{k}'}(t') \right). \tag{3}$$

$\Omega_{\boldsymbol{k}}^{XX}$ includes the instantaneous mean-field XX interaction term (V_{XX}) plus a non instantaneous term originating from 4-particle correlations. The analytical expression of $F(\tau)$ in terms of the two-exciton wavefunctions can be found in Refs. [15, 17–19]. The strong exciton-photon coupling does not modify the memory kernel $F(\tau)$ as a consequence of the fact that 4-particle correlations do not couple to cavity photons [16, 17, 19]. However cavity effects are able to alter the phase dynamics of 2-particle polarization waves $P_{\boldsymbol{k}}$ during collisions, i.e. on a timescale shorter than the decay time of the memory kernel $F(\tau)$. In particular the phase of 2-particle polarization waves $P_{\boldsymbol{k}}$ in SMCs oscillates with a frequency ω_k (fixed by the polariton dispersion relations) modified respect to that of excitons in bare QWs. This fact produces a modification of the integral in Eq. (3). In this way the exciton-photon coupling V affects the XX collisions that govern the polariton amplification process. This mechanism can be more clearly understood simplifying the integral in Eq. (3). Let us consider a situation where the pump, the signal and idler energies are all close to the corresponding polariton resonance values and the broadening are small compared to the polariton splitting $2V$. Then it is a good approximation to replace the integral in Eq. (3) with a simpler expression, adopting the Weisskopf–Wigner [12, 17] approximation used to analyze the spontaneous emission associated to atomic transitions. Within this approximation, the dominant XX interaction term for the signal (0) and idler ($2\boldsymbol{k}$) modes can be written as [17]

$$\Omega_{0(2\boldsymbol{k})}^{XX} = 2\mathcal{F}(\omega_{0(2\boldsymbol{k})}+\omega_{\boldsymbol{k}})\, |P_{\boldsymbol{k}}(t)|^2\, P_{0(2\boldsymbol{k})}(t) + \mathcal{F}(2\omega_{\boldsymbol{k}})\, P_{2\boldsymbol{k}(0)}^*(t)\, P_{\boldsymbol{k}}^2(t) \tag{4}$$

where the complex quantity $\mathcal{F}(\omega)$ is the Fourier transform of the memory kernel $F(\tau)$ plus the mean-field contribution V_{XX}. The first term produces a blue-shift of the polariton resonance and introduces an intensity-dependent dephasing mechanism. The second term provides the coupling mechanism able to transfer polaritons from the pump to the signal and idler modes. An analogous expression can be derived for the XX interaction of the pump mode. This Weisskopf–Wigner description of 4-particle correlation effects in SMCs sets out the relevance of polariton pairs in the scattering process. The energy of these pairs determines the effective scattering rates as shown by Eq. (4) and by the corresponding Feynman diagrams (panel b of Fig. 1). The spectral function $\mathcal{F}(\omega)$ can be calculated by numerical diagonalization of the semiconductor Hamiltonian in the 4-particle subspace. We calculated it for QW excitons following a recent microscopic approach [18, 19] based on the T-matrix (Fig. 1a). The obtained 4-particle spectral density displays strong variations within the spectral region of interest around $2\omega_0$. In particular, moving towards the low energy region, the dispersive part Re (F) increases while the absorptive part that contrasts gain goes to zero. We also observe that the energies of the pump, signal, and idler polaritons are lowered with the increase of polariton splitting ($2V$). The increase of V thus modifies the effective collision rates in such a way that favors the amplification process. This analysis explains the large increase of gain observed when maximizing the exciton-photon coupling [11]. The gain curve versus the input pump power, obtained solving numerically the system of integro-differential equations (1) (6 equations two for each addressed mode) for GaAlAs-based samples with splitting $2V_a = 10.6$ meV ($N_{\text{eff}} = 4$) and $2V_b = 15$ meV ($N_{\text{eff}} = 8$) (Fig. 2), fully confirms this analysis. The calculated gain is defined as the total light intensity transmitted in the signal direction divided by the intensity transmitted in the absence of the pump beam. An increase of less then a factor $1/3$ in the polariton splitting produces an increase of the maximum achievable gain at $T = 10$ K (Fig. 2) of more than one order of magnitude in close agreement with experimental results [11]. The power dependence of gain (Fig. 2) shows an almost exponential growth and then saturates at high powers. The saturation of gain is mainly determined by the nonlinear absorption that

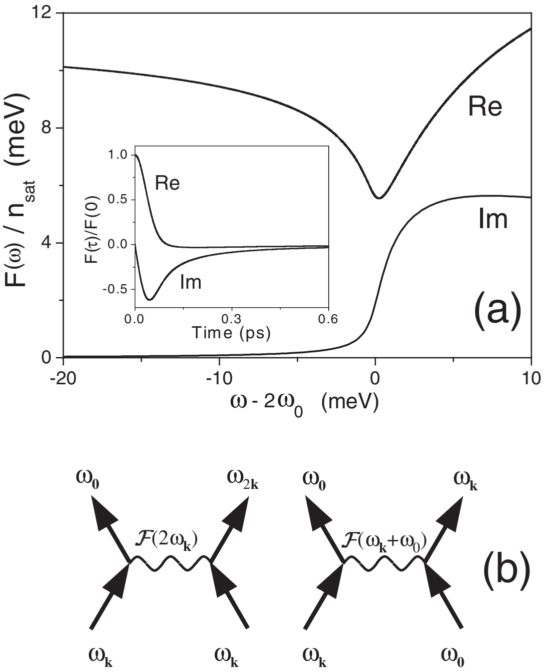

Fig. 1 a) Energy dependence of the effective exciton–exciton scattering potential. It has been calculated for a GaAs QW 7 nm wide with exciton binding energy 13.5 meV and using as dephasing rate of the 4-particle states $\Gamma = 2\gamma_x = 0.58$ meV. The tail of $\mathrm{Im}\,(\mathcal{F}(\omega))$ at negative detuning $(\omega < 2\omega_0)$ is produced by Γ and vanishes for $\Gamma \to 0$. The inset shows the dynamics of the memory kernel $F(\tau)$. b) Feynman diagrams describing the effective exciton–exciton interaction as modified by polariton effects.

is most relevant for the idler beam (according to Eq. (4) the idler nonlinear absorption is determined by Im $\mathcal{F}(\omega_{2k} + \omega_k)$). We observe that the increase of the idler nonlinear absorbance is highly superlinear because the increase of the pump power produces both a direct increase of the exciton density and an increase of Im $\mathcal{F}(\omega_{2k} + \omega_k)$ as a consequence of the blue shift of the polariton-pair resonance $\omega_{2k} + \omega_k$ induced by Re $\mathcal{F}$. We observe that mean-field calculations (also reported in Fig. 2) largely overestimate the experimentally observed gain at pretty high densities.

The results here shown clarifies the origin of the great enhancement of parametric gain observed when increasing the polariton splitting. They also demonstrate that exciton–exciton collisions in semiconductors can be controlled and engineered (e.g. by modifying the Rabi-splitting qnd/or acting on the cavity-exciton detuning). Moreover they give precise indications to produce almost decoherence-free collisions for the realization of all-optical microscopic switches and amplifiers. We believe that the availability of decoherence-free collisions will be crucial for the quantum control and manipulation of the polariton wavefunction inside the cavity and for the realization of microscopic sources of nonclassical light.

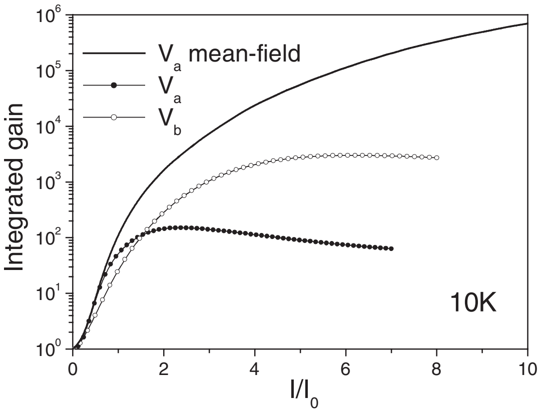

Fig. 2 Power dependence of the integrated gain calculated for two GaAlAs based samples with splitting $2V_a = 10.6$ meV and $2V_b = 15$ meV. $I_0 = 10^{13}$ photons per cm^2 per pulse. The exciting light pulses have 250 fs duration. Material parameters of the two samples are coincident except for N_{eff}. The decay rate of cavity photons through the mirrors $\gamma_c = 0.25$ meV is that of GaAlAs structures considered in Ref. [11].

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